

## Speckle Diffraction of Light by Twinned Orthorhombic Domains: A Sensitive Probe of Interface Topology

K. A. Reza, D. R. Taylor, and R. J. Gooding

*Department of Physics, Queen's University, Kingston, Canada K7L 3N6*

(Received 1 July 1993)

Many compounds have twinned domains with plane walls sufficiently regular to allow Bragg diffraction of light. For a coherent light source we have shown that diffraction peaks in the twinned orthorhombic phase of  $\text{DyVO}_4$  and related compounds have speckle features arising from randomness in domain topology. They evolve with changes in temperature and ordering field, and show qualitative differences between pure  $\text{DyVO}_4$ , where the interfaces are expected to be relatively smooth, and  $\text{Dy}(\text{As}_{0.154}\text{V}_{0.846})\text{O}_4$ , where random strains are expected to roughen and pin the interfaces.

PACS numbers: 61.72.Mn, 42.30.Ms, 75.60.Ch

Many crystalline compounds adopt a twinned lattice configuration, as for example, the twinned orthorhombic domains seen in cuprate superconductors and ferroelectrics such as KDP. In some cases the twinned domains have micron-scale periodicity sufficiently uniform that optical diffraction effects are readily observed [1-3]. Recently we reported the observation of Bragg-regime diffraction of light (i.e., volume as opposed to plane diffraction) in the orthorhombic phase of some rare-earth vanadates and arsenates [4]. The optical diffraction peaks occur at Bragg angles near  $8.0^\circ$  for  $\text{DyVO}_4$  and  $\text{DyAsO}_4$ , and are relatively broad, most likely because of a distribution in the spacings of the  $\{110\}$  planes.

The use of an optical source in these diffraction experiments [4] raises the possibility of observing speckle effects [5] if the scattered light has random phase shifts due to inhomogeneities of the scattering medium. In x-ray or neutron diffraction experiments, the source usually lacks the coherence required [5] for observation of speckle phenomena, although with high-brightness synchrotron x-ray sources the possibility of obtaining a sufficiently coherent source has been demonstrated [6]. Our early experiments [4] could not detect speckle features since we used a photomultiplier detector with a relatively large aperture. In our current experiments we make use of a photodiode array detector that clearly reveals the speckle nature of these diffraction phenomena. The increased sensitivity and information content of speckle diffraction experiments thus provide an important new technique for the study of domain wall properties. In this Letter we describe the main features of speckle diffraction from the twinned orthorhombic phase of  $\text{DyVO}_4$  and  $\text{Dy}(\text{As}_x\text{V}_{1-x})\text{O}_4$ , where  $x=0.154$ , including their dependence on temperature and ordering field. Of particular interest are differences in the speckle features between the pure and mixed compounds attributable to effects of the random strain fields on the domain walls in the mixed compound.

The tetragonal-to-orthorhombic phase transition in  $\text{DyVO}_4$  near 14 K originates in the Jahn-Teller coupling between Dy electronic levels and lattice distortions [7,8].

The two possible orientations of the orthorhombic distortions give rise to a twinned phase with  $\{110\}$  domain walls that can be observed by optical microscopy [9,10]. Samples can be converted to a single domain by application of a stress, or more conveniently, by a magnetic field. The separation and width of the domain walls in such twinned systems depend upon the elastic properties of the lattice. Typically the domain walls have spacings on the scale of a micron and extend through a significant fraction of the sample. In the mixed  $\text{Dy}(\text{As}_x\text{V}_{1-x})\text{O}_4$  crystals, the As-V size mismatch generates random strain fields that are expected to roughen and pin the domain walls [11,12]. Significant differences in domain wall properties are therefore expected between the pure and mixed compounds.

The sample preparation and mounting were the same as described previously. The incident light beam (He-Ne laser at 633 nm) lay in the (horizontal)  $a$ - $c$  plane, a few degrees from the  $c$  axis. The transmitted and diffracted beams could be observed directly on a screen to facilitate alignment. For data collection a Hamamatsu S3901-512Q array detector with 512 photodiode elements was inserted into the diffracted beam and interfaced to a microcomputer for control and data storage. Ordinarily the laser beam was not focused, so that for a typical sample thickness of 1 mm the scattering volume is about  $1\text{ mm}^3$ . Figure 1 shows a typical peak for  $\text{DyVO}_4$  at a temperature well below the transition temperature  $T_D = 14\text{ K}$ . What is plotted in this and other figures is detector output (in arbitrary units) for a *fixed* incident angle  $\theta$  that corresponds to the location of the observed peak. As previously noted [4], diffraction is observed for values of  $\theta$  from almost zero up to  $15^\circ$ , but most of our data are taken for  $\theta$  near  $8^\circ$ .

In almost all cases the diffraction peaks observed with a laser source showed a marked speckle-type structure. The typical angular size of a speckle is approximately 2 mrad, in reasonable agreement with the angle  $\lambda/D \simeq 1$  mrad expected from simple coherence arguments:  $\lambda/D$  represents the angular spread of the scattered light within which the Huygens wavelets from points separated by

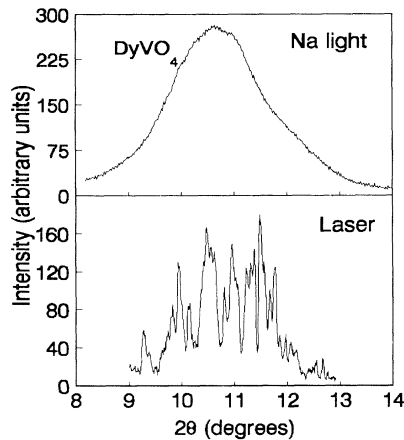


FIG. 1. Comparison of a typical optical diffraction peak in  $\text{DyVO}_4$  at  $T = 10.4$  K for sodium lamp and laser sources showing speckle features in the latter case.

the source of width  $D$  are in phase. In some cases (not shown) we reduced  $D$  by focusing the incident laser beam: the speckle width increased as predicted. As shown in Fig. 1 the speckle effect is entirely absent when the laser source is replaced by a light source with poor coherence (sodium lamp). These observations clearly confirm the speckle effect as the origin of the observed structure in the diffraction peaks. Before discussing the source of the speckle in this situation, we review their qualitative experimental features in the pure and mixed crystals under study.

The temperature dependence of the speckle pattern is of particular interest since it will help to reveal how the topology of the domains develops and evolves below the transition temperature. Figures 2 and 3 show the diffraction peaks for the pure and mixed compounds at several intermediate temperatures and for other incident angles, and will be reported elsewhere. The data allow a number of observations to be made immediately: (1) Deep speckle patterns are the rule for both the pure and mixed samples, implying substantial disorder in domain topology even in pure  $\text{DyVO}_4$ . (2) The speckle features evolve as the temperature is reduced, and of course the intensity builds up strongly (as the order parameter squared [4,12]). In general the speckle pattern continues to change and broaden in  $\text{DyVO}_4$  to quite low temperatures; in the mixed sample the pattern established near  $T_D$  persists to low temperatures with little change. The angular dependence data (not shown) indicate that the peak position of the Bragg diffraction, and hence the mean domain width, do not depend on temperature. (3) A striking difference between the speckle features in the pure and mixed compounds in Figs. 2 and 3 is that in  $\text{DyVO}_4$  there is a narrow temperature range just below  $T_D$  where the diffraction peak does not show speckle, while in the mixed sample, speckle is present as soon as the signal appears out of the

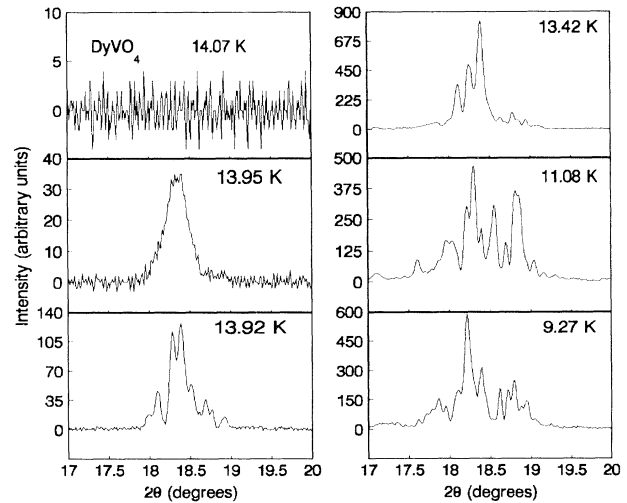


FIG. 2. Evolution of diffraction peak in  $\text{DyVO}_4$  as temperature is reduced through the transition temperature (14.0 K).

noise. (4) For repeated cycling through the transition to the same temperature, the speckle patterns (not shown) for the mixed sample tend to be very similar, while for the pure sample they vary considerably.

Application of an ordering field will directly modify the domain configuration and ultimately produce a single domain. Figure 4 shows the evolution of the diffraction peak in  $\text{DyVO}_4$  for increasing and decreasing ordering field. Initially the diffracted beam intensity increases because the order parameter increases, but then disappears as the domains disappear. When the ordering field is reduced, a twinned domain configuration reappears that is initially quite similar to what was seen at that same

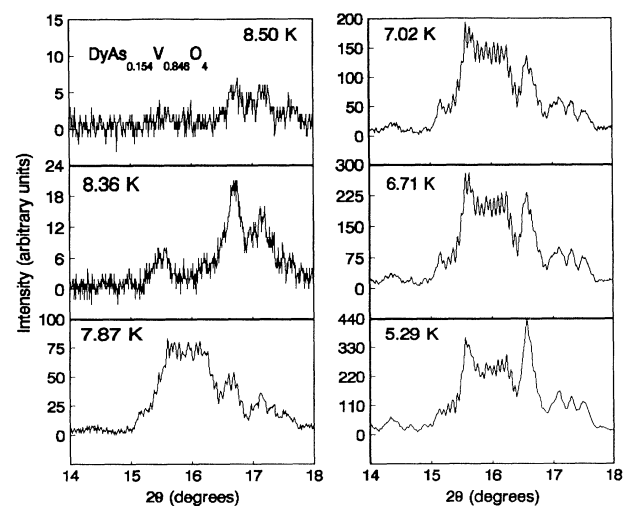


FIG. 3. Evolution of diffraction peak in  $\text{Dy}(\text{As}_{0.154}\text{V}_{0.846})\text{O}_4$  as temperature is reduced through the transition temperature (8.5 K).

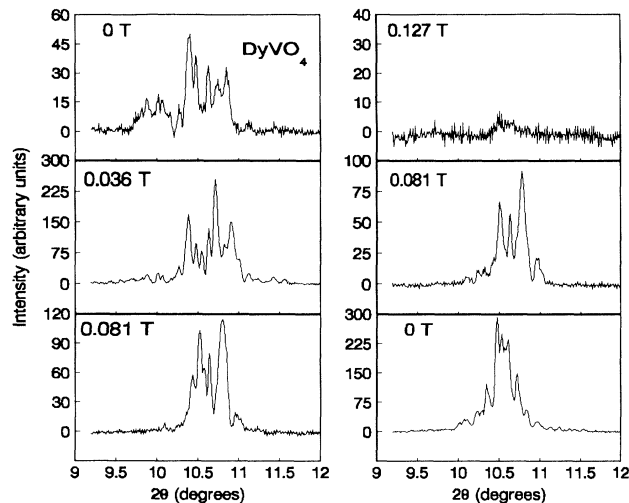


FIG. 4. Evolution of diffraction peak in  $\text{DyVO}_4$  for increasing and decreasing ordering field at fixed temperature (13.5 K).

field when the field was increasing, but at zero field the speckle pattern is very different. Hysteresis of this sort is a feature of domain properties in many systems, and becomes much more prominent as the temperature is reduced. The corresponding data for the mixed sample (not shown) are essentially similar.

There are several types of domain disorder that might be relevant for this system. In a pure crystal with idealized boundaries the domain walls should be equally spaced [13], but the presence of both internal and surface defects, and the influence of realistic surface geometries, will lead to a distribution of domain widths. That a rather broad distribution exists is evident from the range of angles over which Bragg scattering is observed. This randomness in domain widths is a potential source of speckle since the phases of light scattered from each interface will sum randomly at the detector. The various defects that are present in all crystals are expected to be important influences on domain topology and on hysteresis effects [14]. In our system we are dealing with single crystals of good optical quality, but lattice defects as well as chemical impurities are undoubtedly present in significant concentrations. In our mixed samples, substitutional impurities are introduced at high concentrations. While in a pure crystal the elastic forces favor a flat interface, defects present in real crystals will in general induce a rough interface that minimizes the total strain energy. Domain wall roughness is a likely source of speckle, provided that the amplitude of the roughness is a significant fraction of the optical wavelength, and thus of the domain width itself (several percent, say). A third possibility is that the interfaces remain relatively smooth, but are reduced in lateral dimension to adapt to defect stresses that randomly favor one domain or the other throughout the crystal. Some early photographs of the pure system show

that the domain walls are long, comparable to the sample size, in the  $a$ - $b$  plane [9,10], but no comparable information is available for the dimension along the  $c$  axis, nor for domain dimensions in mixed crystals. It should be noted that neutron diffraction data show that the mixed crystals have broader diffraction linewidths than pure crystals below  $T_D$ , implying a greater degree of disorder in the latter case [11] without, however, identifying directly the nature of the disorder.

It would be desirable to reconstruct the domain topology in the scattering volume by analysis of the observed speckle patterns, but this is not possible in practice. An alternative approach, which we are pursuing, is to compute the speckle pattern from model domain configurations and look for patterns of behavior similar to those observed—these calculations are straightforward, as described, e.g., in Ref. [15]. As a first step, we have carried out simulations for a pseudo-one-dimensional model of the domain topology. For a linear array of (flat) domain walls there are two different structural domains that may be distinguished by their strains away from the tetragonal phase. Thus, we have “grown” a computer-generated crystal with atomic positions given by  $\{x_i\}$ , and the crystal structure produced corresponds to a distribution of domains with alternating orthorhombic strain states. The domain sizes are randomly assigned, having a fixed mean value (i.e., an unweighted distribution of domain sizes having a lower and upper bound was used), and then the static structure factor, viz.  $S(q) = (1/N) |\sum_i \exp(iqx_i)|^2$ , is found for a particular specification of domain sizes. A similar scheme has been used to study stacking faults [16]. (This approach is consistent with one additional experimental result: an aperture was used to allow differing subvolumes of the crystal to serve as the scattering volume at one fixed temperature. We found that the obtained speckle patterns had no similarity to one another; viz., for a given quenched domain wall disorder, a given speckle pattern is found.) The magnitude of the orthorhombic strains were then allowed to increase with decreasing temperature according to the observed [12] critical exponent  $\beta = 0.3$ : to be specific, for  $e_O$  being the magnitude of the orthorhombic strain, one has  $e_O(t) \propto \pm(-t)^{0.3}$ , where  $t$  is the reduced temperature. We have found that the shape of the interface connecting a pair of domains does not affect the speckle pattern (provided the interfaces are flat) and have therefore employed the strain soliton predicted in Ref. [13].

An example of a diffraction peak computed for this model is shown in Fig. 5. There the domain sizes  $L$  are randomly distributed in the range  $\Delta L = \pm 10\%$  about a mean value  $\bar{L} = 200$  layers (the observed scattering profile is sensitive to this spread in domain-wall spacings—the angular breadth of the speckle pattern does expand when the ratio  $\Delta L/\bar{L}$  is increased). The scattering volume has a dimension corresponding to 800000 atomic layers; with this many layers we found no sig-

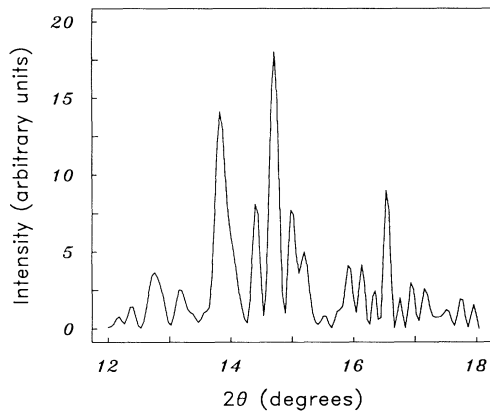


FIG. 5. Example of a computed speckle diffraction peak for a temperature  $0.75T_D$  and a distribution of domain widths that is 10% of the average wall separation.

nificant finite size effects in the structure factor. Each disordered crystal gives rise to a unique scattering profile (consistent with the experimental observation mentioned above), and our goal is to recognize trends in the speckle patterns, and associate them with microscopic aspects of the domains. The computed profile demonstrates that a speckle pattern comparable to those observed can be obtained from this simple model. Further, when studying the manner in which such computer generated patterns change, one trend consistent with experiment was observed: As the temperature is decreased the computed integrated intensity increases without a change in the speckle pattern, similar to Fig. 3, and thus we infer that there is strong pinning of the domain wall locations for the doped crystal.

It is this kind of qualitative information that we feel can be obtained from a combination of experiment and numerical modeling. It seems clear, however, that more realistic treatments that allow for roughness (something that can only be considered in a fully two-dimensional model) and thermally activated depinning of domain walls will be required to explain the differences between the behaviors of the pure and mixed compounds, as our preliminary numerical studies have suggested [17].

The temperature data of Figs. 2 and 3 and our simple theoretical model show that in both the pure and mixed crystals there is sufficient domain disorder to create a speckle pattern, but the As impurities present in the mixed crystal are effective in establishing domain structures and pinning them against temperature evolution. The lack of speckle observed in  $\text{DyVO}_4$  just below  $T_D$  is very interesting, and implies that the domain walls are relatively smooth at that temperature. Significant domain wall motion may be present at this temperature, since for temperatures just below  $T_D$  the effective mass of a wall (which grows with the order parameter squared) will be very small, thus making the walls quite glissile. However, our attempts to detect frequency shifts in the scattered light by a light-beating experiment were un-

successful.

In considering the ordering field results, it should be recalled that in the basic models of the influence of a field on domains, the favored domains simply grow fatter while the disfavored grow thinner [14]. Hence the "repeat distance" of the domain structure should not change, and this was confirmed in experiments that showed that the angular position of peak Bragg diffraction did not change during runs such as that shown in Fig. 4. An important result indicated in Fig. 4 is that the final domain configuration reached after application and removal of the ordering field is, on the basis of the speckle pattern, apparently smoother and more regular than the original domain configuration.

In summary, we have demonstrated a simple optical technique that in suitable (transparent) systems reveals domain structure on length scales of hundreds of Å. Our results show the capability of distinguishing between structures induced by thermal fluctuations and by static disorder. The variation of the speckle pattern with decreasing temperature, and when the system is cycled through  $T_D$  several times, is most likely associated with changing thermodynamic restoring forces and weak pinning centers, and this idea is being explored theoretically.

We acknowledge valuable discussions with M. Sutton and A. Cairns, and technical assistance from R. Noteboom and S. Gillen. Research support was provided by NSERC of Canada and by Queen's University.

- [1] W. Kleemann and J. Ferré, *Phys. Rev. B* **24**, 1568 (1981).
- [2] T. Tsukamoto, J. Hatano, and H. Futama, *J. Phys. Soc. Jpn.* **51**, 3948 (1982).
- [3] R. Gagnon, M. Oussena, and M. Aubin, *J. Cryst. Growth* **121**, 559 (1992).
- [4] K.A. Reza and D.R. Taylor, *J. Phys. Condens. Matter* **3**, 7533 (1991).
- [5] M. Francon, *Laser Speckle and Applications in Optics* (Academic, New York, 1979).
- [6] M. Sutton *et al.*, *Nature (London)* **352**, 608 (1991).
- [7] G.A. Gehring and K.A. Gehring, *Rep. Prog. Phys.* **38**, 1 (1975).
- [8] A. Kasten, *Z. Phys. B* **38**, 65 (1980).
- [9] K.A. Gehring and M. Rosenberg, *Phys. Status Solidi b* **47**, K75 (1971).
- [10] L. Klein, H.G. Kahle, H.C. Schopper, and H. Walter, *Int. J. Magn.* **3**, 17 (1972).
- [11] J.T. Graham, D.R. Taylor, D.R. Noakes, and W.J.L. Buyers, *Phys. Rev. B* **43**, 3778 (1991).
- [12] K.A. Reza and D.R. Taylor, *Phys. Rev. B* **46**, 11425 (1992).
- [13] A.E. Jacobs, *Phys. Rev. B* **46**, 8080 (1992).
- [14] C. Kittel and J.K. Galt, in *Solid State Physics*, edited by H. Ehrenreich and D. Turnbull (Academic, New York, 1956), Vol. 3, p. 437.
- [15] A. Garg and D. Levine, *Phys. Rev. Lett.* **60**, 2160 (1988).
- [16] R. Berliner and R.J. Gooding, *Acta Crystallogr. A* (to be published).
- [17] A. Cairns, R.J. Gooding, and D.R. Taylor (unpublished).